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(54) Mixture of long glass fiber-reinforced polypropylene and polypropylene resin and moldings formed therefrom

Mischung aus mit langen Glasfasern verstärktem Polypropylen und Polypropylenharz und daraus hergestellte Formteile

Mélange de polypropylène renforcé de fibres de verre longue, et de résine de polypropylène et pièces moulées obtenues à partir de celui-ci

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(73) Proprietor: Kawasaki Steel Corporation
Chuo-ku, Kobe-shi, Hyogo 651 (JP)

(72) Inventor: Yoshino, Kenji,
c/o Kawasaki Steel Corporation
Chiyoda-ku, Tokyo 100 (JP)

(74) Representative: Overbury, Richard Douglas et al
Haseltine Lake & Co,
Imperial House,
15-19 Kingsway
London WC2B 6UD (GB)

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Description

[0001] The present invention relates to a molding mixture for forming a long glass fiber reinforced polypropylene molding having high mechanical strength and to a molding formed of the molding mixture.

Description of the Related Art

[0002] Glass fiber reinforced polypropylene resins have been widely used for various industrial product components because they possess excellent mechanical strength, heat resistance and formability. Long fiber reinforced polypropylene resins obtained by a drawing process in which continuous fibers are impregnated with a resin while being drawn exhibit superior impact strength, creep characteristics and vibration fatigue characteristics over short fiber reinforced polypropylene resins obtained by kneading chopped strands and a resin with an extruder or molding machine. Long fiber reinforced polypropylene resins produced by a drawing method also enables the binding of glass fibers at a very high density, because the glass fibers extend in a single direction in a bound state.

[0003] It is difficult to produce a short fiber reinforced polypropylene resin by kneading the resin with an extruder if the glass fiber content exceeds 50 % by weight. On the other hand, long glass fiber reinforced polypropylene resin is easily produced by a drawing method even if the glass fiber content is 80% or higher by weight. Due to this advantage, long fiber reinforced polypropylene resins produced by drawing and having a large glass fiber content are frequently utilized as a master batch. When long fiber reinforced polypropylene resin possessing large glass fiber content is used as a master batch, the range from which the proportion of a diluting polypropylene mixed in the long fiber reinforced polypropylene resin is selected increases and, accordingly, melt process moldings of long fiber reinforced polypropylene having various fiber contents which are suitable for various kinds of use or purposes can be obtained. Thus, the use of long fiber reinforced polypropylene resins is economical because of their wide product application range.

[0004] However, long fiber reinforced polypropylene resins produced by drawing to be used as a master batch have drawbacks in that it is difficult to completely impregnate several thousands or several tens of thousands of glass fiber filaments with a polypropylene resin while drawing the filaments. Another drawback is that hydrophilic glass fibers have low wettability with non-polar polypropylene resins. Methods for reducing these drawbacks have been disclosed, as described below.

[0005] For example, Japanese Patent Laid-Open Publication 17631/1993 discloses a method of using a low-viscosity polypropylene of a melt flow rate of 30 g/10 min or higher measured by ASTM method D-1238 (load: 2.16 kg, temperature: 230°C) to facilitate molten resin impregnation, Japanese Patent Publication No. 25340/1991 discloses a method of impregnating glass fibers with a resin having a very small molecular weight, and Japanese Patent Laid-Open Publication No. 181528/1991 discloses a method of finishing the surface of glass fibers and using a modified polypropylene to improve the wettability.

[0006] Methods for obtaining moldings by a master batch process have also been disclosed, as described below.

[0007] Japanese Patent Laid-Open Publication No. 241406/1989 discloses a method of molding a dry blend of a long fiber reinforced thermoplastic resin and a thermoplastic resin having no long fibers, Japanese Patent Laid-Open Publication No. 114705/1991 discloses a method of using a polypropylene resin of a melt flow rate of 20 to 60 g/10 min or higher and pellets having a small size, i.e., a shape in which the length of at least one side is 1 mm or less and the specific surface area is 20 cm²/g or larger, and Japanese patent publication No. 20339/1984 discloses a method of heating a combined filament yarn fabric of glass fibre and modified polypropylene fibre.

[0008] Japanese patent publication No. 5239286 discloses resin compositions re-inforced with glass fibres. The compositions comprise 10 to 70 wt% of a mixture of a propylene homopolymer or copolymer which has a melt index of at least 300g/10min and glass fibre, and 90 to 30 wt% of a propylene/ethylene copolymer. The resin composition does not contain any finishing agent for the glass fibre and has a disadvantageously high melt index value.

[0009] Japanese patent publication No. 62086026 discloses a sizing composition for glass fibres which comprises a combination of a silane coupling agent having a defined formula and a zirco-aluminate coupling agent having a defined formula. The sizing agent can be coated on to glass fibres to enhance fluidity.

[0010] However, in prior art processes using a long glass fiber reinforced polypropylene resin as a master batch, a molding obtained by diluting such a resin and by molding the mixture tends to decrease in strength as glass fiber content increases. This tendency intensifies when the glass fiber content exceeds about 60% by weight.

[0011] The above-described prior art has been discussed with respect to impregnation techniques, particularly, methods for increasing fiber content, improving wettability, and obtaining moldings from master batches.

[0012] However, the above-described problems experienced when using a long glass fiber reinforced polypropylene resin as a master batch have not been addressed by the prior art and thus no method for solving the problem has been disclosed. If the problem of strength reduction in a molding obtained by using such a master batch is solved, moldings of long glass fiber reinforced polypropylene resins which satisfy the requirements for a wide array of uses and purposes could be produced at a low cost.

SUMMARY OF THE INVENTION

[0013] According to one aspect of the present invention, there is provided a mixture for producing melt process moldings, the mixture comprising: from 5 to 70 wt% of a master batch of pellets comprising continuous reinforcement glass fibres, the pellets having a length of 2 to 50 mm in the direction along the fibres and the glass fibres in the pellets extending uniformly in parallel with each other through a distance substantially equal to the length of the pellets, and from 30 to 95 wt% of polypropylene resin, characterised in that the glass fibres are surface-finished with a finishing agent containing a coupling agent and impregnated with a modified polypropylene resin bonded to the coupling agent and having a melt flow rate of from 70g/10min to less than 300 g/10min (ASTM D-1238, load: 2.16 kg, temperature: 230°C).

[0014] According to a second aspect of the present invention, there is provided a method of preparing a mixture for producing melt process moldings comprising: forming glass fibres into a bundle; impregnating the bundle with a modified polypropylene resin having a functional group capable of chemically bonding with a coupling agent; dividing the resulting impregnated bundle into a multiplicity of pellets; and mixing the pellets with polypropylene resin, characterised in that the glass fibres are coated with a finishing agent containing a coupling agent before they are formed into a bundle, and in that the modified polypropylene resin has a melt flow rate of from 70g/10min to less than 300g/10min (ASTM D-1238, load : 2.16 kg, temperature 230°C).

[0015] According to a third aspect of the present invention, there is provided a method of preparing a mixture for producing melt process moldings comprising: impregnating a bundle of glass fibres with a modified polypropylene resin having a functional group capable of chemically bonding with a coupling agent; dividing the resulting impregnated bundle into a multiplicity of pellets; and mixing the pellets with polypropylene resin, characterised in that the method includes the step of coating individual glass fibres with a finishing agent containing a coupling agent, and in that the modified polypropylene resin has a melt flow rate of from 70g/10min to less than 300g/10min (ASTM D-1238, load : 2.16 kg, temperature 230°C).

[0016] In order to solve the above-described problem, the inventors of the present invention conducted various studies of melt process molding mixtures and moldings subsequently formed, each mixture consisting of a long glass fiber reinforced polypropylene master batch and a polypropylene resin used to form a high-strength molding. From the studies, the factors found to be most vital to success are:

- uniform dispersion of glass fibers in a master batch produced by drawing;
- sufficiently high adhesion of interface between glass fibers and a polypropylene resin in a master batch; and
- maintaining matrix resin viscosity in a master batch within a particular range to reduce the apparent viscosity of the entire master batch and to ensure the desired strength.

[0017] Based on the above-described findings, the inventors found that a melt process molding formed of a mixture of a long glass fiber reinforced polypropylene resin master batch and a polypropylene resin can be obtained with suitable glass fiber dispersion, excellent surface appearance and outstanding strength only if the glass fiber and the resins are prepared under particular conditions.

[0018] In preparing a long glass fiber reinforced polypropylene having high mechanical strength, a polypropylene of high molecular weight and strength, in other words, having a low melt flow rate, is preferred as a matrix even if wettability and adhesion are improved by finishing the surface of glass fibers and by using a chemical bond formed by a modification of polypropylene as described above.

[0019] On the other hand, for uniform dispersion of long glass fibers as a reinforcement member in a matrix, it is necessary to impregnate a bundle of glass fibers consisting of several thousands to several tens of thousands of filaments with polypropylene. In such a case, a polypropylene having high fluidity, i.e., a high melt flow rate, is more preferable because of its high permeability. Thus, there is a dichotomy with respect to matrix polypropylenes.

[0020] Japanese Patent Laid-Open No. 114705/1991 discloses a master batch in which a polypropylene having very high fluidity in comparison with the conventional matrix polypropylene, i.e., a melt flow rate of 60 g/10 min (20 to 60 g/10 min), is used as a matrix to achieve a large glass fiber content, and which is formed into a plate-like shape for improved heat conductivity. In the present technical field, it is commonly believed that in using such a highly fluid master batch matrix polypropylene resin, strength is sacrificed.

[0021] The inventors conducted an experiment in which, as described below with respect to Comparative Example 4, a master batch possessing a large fiber content and produced in the shape of pellets was drawn using a polypropylene having a melt flow rate of 60 g/10 min as a matrix, mixed with a diluting polypropylene and thereafter melted and molded. It was confirmed that even in this master batch the apparent viscosity was very high and the glass fibers were not easily dispersed through the entire resin.

[0022] However, in the course of studying the present invention, the inventors produced a master batch having a large glass fiber content by using as a matrix polypropylene having very low resin strength and markedly high fluidity

in comparison with the conventional master batch matrix polypropylene, i.e., a melt flow rate of from 70 to 300 g/10 min. The strength of this master batch itself was low. However, the inventors also made a melt process molding by mixing this master batch and a diluting polypropylene, and discovered that the strength of this melt process molding was surprisingly larger than that of a conventional melt process molding obtained from a master batch in which a low melt flow rate polypropylene is used.

[0023] The present invention provides a mixture for melt process moldings consisting of a long glass fiber reinforced polypropylene resin and another polypropylene resin, and a molding formed of the mixture, the mixture being prepared in such a manner that a bundle of continuous reinforcement glass fibers surface-finished with a finishing agent containing a coupling agent is impregnated with a modified polypropylene resin having a functional group capable of chemically bonding with the coupling agent and having a melt flow rate of from 70 to 300 g/10 min (ASTM D-1238, load: 2.16 kg, temperature: 230°C) while the bundle of glass fibers is being drawn. The bundle of glass fibers is cut after the impregnation to form pellets (as a master batch) which have a length of from 2 to 50 mm in the direction along the fibers, in which the glass fibers uniformly extend in parallel with each other through a distance substantially equal to the length of the pellet, and the mixture being formed of from 5 to 70 wt% of the master batch and from 30 to 95 wt% of a polypropylene resin.

[0024] The above-described advantages as well as other features and advantages of the present invention will become apparent from the following detailed description of the present invention along with selected variations of the invention.

DETAILED DESCRIPTION OF THE PREFERRED EMBODIMENTS

[0025] Embodiments of the present invention will hereinafter be described in detail with reference to specific forms of the invention, but specific terms used in the specification are not intended to limit the scope of the invention which is described in the appended claims.

[0026] Glass fibers used in accordance with the present invention are fibers of E-glass, S-glass, C-glass, AR-glass, T-glass, D-glass, R-glass or the like. Ordinarily, fibers of such glass are provided as a twisted bundle of a plurality of glass filaments, i.e., as a glass roving. Glass fibers having a diameter of from 3 to about 40 μm are suitable. If the glass fiber diameter is smaller than 3 μm , and if the glass content is fixed, the number of glass fibers is increased such that it is difficult to impregnate the glass fiber bundles with a resin. If the glass fiber diameter is larger than 40 μm , a considerable deterioration results in the external appearance of a molding. Glass fibers having a diameter within the range of from 9 to 20 μm are most suitable.

[0027] Glass fibers used in accordance with the present invention must be surface-finished with a finishing agent containing a coupling agent. Non-surface-finished glass fibers have a hydrophilic nature and therefore have low wettability with polypropylene resins having a lipophilic nature. As a result, a large surface tension is caused between such glass fibers and polypropylene, and surface-finished glass fibers cannot easily be impregnated with a polypropylene resin. Consequently, masses of glass fibers unimpregnated with the resin in such a molding cause external appearance defects and strength reduction.

[0028] The coupling agent for surface finishing may be selected from silane coupling agents, such as aminosilane, epoxysilane, amidosilane, azidosilane and acrylsilane, titanate coupling agents, and mixtures of these. Among these materials, aminosilane or epoxysilane, particularly an aminosilane coupling agent is most suitable.

[0029] The finishing agent may also contain components other than the described coupling agents, for example, a binder, which is, preferably, an urethane resin, an epoxy resin, a vinyl acetate resin, or a modified polyolefin or non-modified polyolefin, such as polyethylene or polypropylene modified with (meth)acrylic acid or acid anhydride, and a lubricant, such as a cationic, nonionic, anionic or silicon compound. Preferably, the content of the finishing agent in the glass fibers is from 0.05 to 3 wt%. If the content of the finishing agent is smaller than 0.05 wt%, the surface finishing effect is inadequate. If the content of the finishing agent exceeds 3 wt%, undesirable coloring and a deterioration due to heat can occur in a molding.

[0030] A modified polypropylene resin having a functional group capable of chemically bonding with the above-described coupling agent is preferably selected from carboxylic acid-modified polypropylene resins, acid anhydride-modified polypropylene resins and epoxy-modified polypropylene resins. These polypropylene resins can be obtained by melt-kneading a monomer of (meth)acrylic acid, maleic acid, itaconic acid, maleic anhydride, citraconic anhydride, itaconic anhydride (meth)acrylic acid glycidyl ester with a polypropylene resin under the presence of a radical initiator in an extruder.

[0031] The content of the functional group capable of bonding with the coupling agent in the modified polypropylene resin is, preferably, from 0.01 to 5 wt% in terms of the content of a modification monomer. If the content of the functional group is smaller than 0.01 wt%, the amount of polymer grafted on the glass surface is insufficient, resulting in a reduction in the strength of a molding. If the content of the functional group is larger than 5 wt%, coloring, i.e., a reduction in heat resistance strength, occurs in the molding. A maleic anhydride-modified polypropylene resin grafted with from 0.02 to

1.5 wt% of maleic anhydride is most suitable.

[0032] The above-described modified polypropylene resin may comprise a single modified polypropylene resin or a mixture of non-modified polypropylene resin and modified polypropylene resin. The mixture of the latter case is also considered a modified polypropylene resin and is included within the scope of the present invention. However, any mixture proportion of polypropylene resins should suffice as long as the total melt flow rate falls within the range of from 70 to 300 g/10 min.

[0033] The modified polypropylene resin comprises a polypropylene resin grafted with a modification monomer, as described above. However, polypropylene resins referred to herein comprise those containing 70 wt% or more of polypropylene in polymer chains. Examples include: propylene homopolymer; and copolymers containing less than 30 wt% of vinyl groups containing monomers copolymerizable with propylene and 70 wt% or more of propylene. Examples of such copolymers are propylene-ethylene random and block copolymers, propylene-butene copolymers, and propylene-EPDM copolymers.

[0034] One or a mixture of two or more of these modified polypropylene resins may be used. Examples of the modified polypropylene resin most preferred include polypropylene homopolymer modified with from 0.02 to 1.5 wt% of maleic anhydride and an ethylene-propylene copolymer modified with from 0.02 to 1.5 wt% of maleic anhydride and having an ethylene content of 5 wt% or less.

[0035] In the present invention, glass fibers are surface-finished with a finishing agent containing a coupling agent and a modified polypropylene resin is used as an impregnation resin. The polypropylene resin is thereby grafted to the finishing agent on the glass fiber surface so that the strength of the interface between the glass fiber and the polypropylene matrix resin is increased and the strength of a molding formed from these materials is markedly increased.

[0036] It is necessary for the modified polypropylene resin formed as a matrix resin of a master batch in accordance with the present invention to have a melt flow rate in the range of from 70 g/10 min to 300 g/10 min as measured by ASTM method D-1238 (load: 2.16 kg, temperature: 230°C) (all melt flow rates shown below are values measured by the same method). If the melt flow rate is lower than 70 g/10 min, glass fibers are not uniformly dispersed when a mixture of the master batch and a polypropylene resin is melted and molded. As result, non-uniform glass fiber density exists in the molding obtained, and low molding strength results. If the melt flow rate is higher than 300 g/10 min, low molding strength again results because the matrix resin has a low molecular weight, although the glass fibers are suitably dispersed in the molding. An optimal melt flow rate of the modified polypropylene resin is within the range of from 80 to 150 g/10min.

[0037] Master batch pellets of the long glass fiber reinforced polypropylene resin in accordance with the present invention are obtained in such a manner that the above-described continuous reinforcement glass fiber bundles are impregnated with the above-described modified polypropylene resin while being drawn and are cut in the direction perpendicular to the direction in which the fibers are drawn. Any method may be used to impregnate the fiber bundles with the resin. For example, the impregnation method may involve any of the following methods: impregnating the glass fiber bundles with an emulsion of the modified polypropylene resin so that the emulsion is attached to the entire surface of the fibers, and thereafter drying the emulsion; attaching a suspension of the modified polypropylene resin to the glass fibers, drying the suspension and thereafter heat-melt-impregnating the fiber bundles with the resin; charging the glass fibers to attach a powder of the modified polypropylene resin to the glass fibers and thereafter heat-melt-impregnating the fibers with the resin; impregnating the glass fiber bundles with a solvent in which the modified polypropylene resin is dissolved, and thereafter removing the solvent; heating a mixture of the modified polypropylene resin and the continuous glass fibers so that the glass fiber bundles are impregnated with the molten modified polypropylene resin; impregnating the glass fiber bundles with the modified polypropylene resin heated and melted while opening the glass fibers on a bar, a roll and a die; or other methods. Among these methods, the method of impregnating the fiber bundles with the modified polypropylene resin heated and melted while opening the glass fibers on a bar, a roll and a die is most preferable because of the simplicity of the apparatus and process involved.

[0038] In master batch pellets thus formed and cut, the glass fibers extend completely through the pellet length in one direction in parallel with each other. The pellets contain from 60 to 90 wt% of the glass fibers. The pellet length is from 2 to 50 mm in the direction along the fibers. If the glass fiber content is smaller than 60 wt%, the above-mentioned advantages of the master batch are significantly reduced. Furthermore, the product application range of a molding comprising such pellets after dilution is restricted and the industrial value of the product is thereby reduced. If the glass fiber content is higher than 90 wt%, the glass fiber bundles cannot be sufficiently impregnated with the resin and it is difficult to obtain the desired product. The glass fiber content is, preferably, within the range of from 70 to 85 wt%. If the pellet length is smaller than 2 mm, the glass fiber length in the product is so short that the molding strength, particularly the strength against impacts, is reduced. If the pellet length is larger than 50 mm, clogging in the hopper and mixture segregation of the master batch with a polypropylene resin takes place during injection molding or extrusion. The pellet length is, preferably, from 3 to 25 mm. Any pellet shape is acceptable as long as the length is from 2 to 50 mm. For example, the pellets may be circular, elliptic or rectangular in section. Preferably, the longitudinal length of a cross section of the pellets is such that the aspect ratio (the ratio of the pellet length and the cross section length)

is from 1 to 10.

[0039] A diluting polypropylene resin to be mixed with the master batch pellets of the long glass reinforced polypropylene resin used in accordance with the present invention comprises 70 wt% or more of polypropylene as a constituent unit. Examples include: propylene homopolymers, copolymers containing less than 30 wt% of vinyl group containing monomer copolymerizable with propylene and 70 wt% or more of propylene, and mixtures of these. Examples of such copolymers are propylene-ethylene random copolymers, propylene-ethylene block copolymers, propylene-butene copolymers, and propylene-EPDM copolymers. The melt flow rate of these diluting polypropylene resins is not particularly limited. However, melt flow rates lower than 1 g/10 min reduce fluidity to where moldability is disadvantageously reduced. Melt flow rates exceeding 200 g/10 min reduce molding strength after dilution, particularly strength against impacts. Preferably, the melt flow rate is within the range of from 5 to 150 g/10 min.

[0040] Diluting polypropylene resins may be selected according to purposes for which moldings are used. For thin moldings, those having a high fluidity, i.e., a melt flow rate of from 30 to 100 g/10 min, are suitable. If substantially high impact resistance is required, it is preferable to use ethylene-propylene block copolymer having a melt flow rate of from 3 to 20 g/10 min and containing from 10 to 30 wt% of ethylene, or propylene-EPDM copolymer having a melt flow rate of from 3 to 20 g/10 min and containing from 10 to 30 wt% of EPDM.

[0041] If a long fiber reinforced ethylene-propylene block copolymer or propylene-EPDM copolymer molding is produced in a prior art manner, and if high impact resistance is required, impregnation of the glass fibers is difficult because of the high melt viscosity of ethylene-propylene block copolymer or propylene-EPDM copolymer. Accordingly, the present invention method of molding a mixture of the above-described master batch and a like high-impact-resistance copolymer is remarkably effective.

[0042] The weight ratio of the long glass fiber reinforced polypropylene master batch to the diluting polypropylene resin in the mixture in accordance with the present invention is from 5/95 to 70/30. If the proportion of the master batch in the mixture is smaller than 5 wt%, the glass fiber content in a molding after dilution is so small that the high strength/impact resistance advantage of a long fiber reinforced molding is not sufficiently exhibited. If the proportion of the master batch is larger than 70 wt%, that is, if the proportion of the diluting polypropylene resin is smaller than 30 wt%, the economic advantage gained by using the diluting polypropylene is reduced. Furthermore, the product application range of the resulting molding is restricted, so that the industrial value of the molding is reduced. An optimal master batch/polypropylene weight ratio is within the range of from 20/80 to 50/50.

[0043] The shape of the diluting polypropylene is not particularly limited. However, it is preferable to provide the diluting polypropylene in the form of pellets. More preferably, the size and shape of pellets of the diluting polypropylene are close to those of the master batch pellets. If the size of the pellets of the two materials greatly differ from each other, the two materials separate from each other in a hopper during melt molding. Preferably, the polypropylene resin has the shape of grains having a diameter of from 2 to 5 mm or a cylindrical shape having a diameter of from 1 to 5 mm and a length of from 1 to 10 mm.

[0044] Various additives may be added to the mixture for melt process moldings in accordance with the present invention. Examples of such additives are antioxidants, such as phenolic and phosphoric antioxidants, ultraviolet absorbers, dyestuffs or pigments for coloring, higher fatty acid, metallic salts of higher fatty acid, fatty amide, and lubricants such as waxes.

[0045] Examples of melt processes for molding the mixture in accordance with the present invention include an ordinary injection molding machine, an injection-press molding machine, a single screw extruder, a double screw extruder, and a hot pressing machine. Examples of moldings thereby obtained include ordinary injection moldings, rod-like moldings formed by contour extrusion, plate-like moldings formed by contour extrusion, and sheets.

[0046] The average (median length) of the permanent length of fibers in a molding obtained by melting and molding the mixture of the above-described master batch and polypropylene resin is preferred to be from 0.8 to 10 mm.

[0047] If the median length is smaller than 8 mm, the advantages of long fiber reinforced moldings such as high strength, large elastic modulus, improved creep characteristics and improved vibration fatigue characteristics cannot be adequately realized.

[0048] At a median length of 10 mm, the limits of the above-mentioned benefits associated with long fiber reinforced moldings are reached and external appearance deterioration begins to occur.

[0049] In order to maintain the average permanent glass fiber length at from 0.8 to 10 mm, it is preferred that a comparatively moderate melt molding condition be set for the melt process molding mixture of the present invention, i.e., a smaller kneading force which reduces the shearing force applied to the molten resin.

[0050] For example, it is preferred that a deep-root screw be employed to set the back pressure of the screw to a minimum value of from 0 to 3 kg/cm².

Alternatively, a method of reducing the screw revolutions may be used. An advantage of the present invention is that the glass fibers in the melt process molding mixture can be sufficiently dispersed with a reduced kneading force.

[0051] By means of the present invention, high-strength melt process moldings can be obtained for the following reason. The content of long glass fibers in the master batch is large and the adhesion between the glass fibers and

the polypropylene resin in the master batch is improved by surface finishing of the glass fibers and modification of the polypropylene resin. Further, it is thought that the polypropylene of the master batch can disperse easily in the diluting polypropylene because the master batch polypropylene, having strong bonds around the surfaces of the glass fibers and having high fluidity, has an affinity with the diluting polypropylene and has its apparent viscosity reduced at the time of melting and molding.

[0052] Accordingly, it is thought that a molding having a higher strength can be obtained even when the melt flow rate of the master batch polypropylene is high, if the melt flow rate of the diluting polypropylene is reduced.

Examples

[0053] The following examples are merely illustrative and are not intended to define or limit the scope of the invention, which is defined in the appended claims.

(Examples 1 to 5)

[0054] A mixture of 100 parts by weight of polypropylene homopolymer having a melt flow rate of 8 g/10 min, 2 parts by weight of maleic anhydride and 0.3 parts by weight of 2,5-dimethyl-2,5-di(t-butylperoxy)hexyne -3 was melted and kneaded at 180°C and extruded by a bent double screw extruder to obtain a maleic anhydride-modified polypropylene having a melt flow rate of 120 g/10 min and a grafted maleic anhydride content of 1.2 wt%.

[0055] Mixtures of 20 wt% of the modified polypropylene resin obtained in this manner and 80 wt% of polypropylene homopolymers having melt flow rates of 60 g/10 min (Example 1), 80 g/10 min (Example 2), 120 g/10 min (Example 3), 200 g/10 min (Example 4), and 300 g/10 min (Example 5) were melted and kneaded at 220 °C and were poured into an impregnation bath in which five metallic rolls were disposed and which was maintained at a temperature of 280°C. Five bound glass rovings, each of which contained 2320 glass filaments surface-finished with a finishing agent containing 0.2 wt% of γ -aminopropyltriethoxysilane and having a diameter of 16 μm (total number of filaments: 11600) were alternately passed along the five rolls in the molten resin of the impregnation bath, and a tensile force of 2 kg was applied to the rovings, thereby drawing the rovings at a speed of 1 m/min (residence time: 30 seconds). The glass filaments opened on the rolls, thereby impregnating the glass fiber rovings with the resin. The glass fiber rovings impregnated with the resin were passed through a die, cooled and thereafter cut into 10 mm pellets with a cutter. The pellets were incinerated in a muffle furnace at 450°C for 8 hours, and the glass content was measured at 75 wt%. The incinerated pellets contained glass fibers extending parallel to each other having the same length as the pellets. Melt flow rates of molten resin samples from the impregnation bath were measured at 82 g/10 min, 95 g/10 min, 125 g/10 min, 190 g/10 min, and 280 g/10 min.

[0056] The above-obtained master batch pellets, containing 75 wt% of glass fibers, were mixed with homopolymer polypropylene resin pellets having 3 mm diameter grains and a melt flow rate of 45 g/10 min in a respective 53.3 wt%/46.7 wt% ratio, and the mixture was injection-molded by a 50t clamping force injection molding machine. Injection molding was conducted at a temperature of 240°C, with the mold temperature at 60°C. Table 1 shows the results of tensile strength (ASTM D-638) and Izod impact strength (ASTM D-256, notched) measurements taken on the obtained moldings.

Table 1

	Melt Flow Rate of Master Batch Matrix Resin (g/10 min)	40wt% Glass Fiber Reinforced Molding after Dilution	
		Tensile Strength (kg/cm ²)	Izod Impact Strength (kg·cm/cm)
Example 1	82	1140	37
Example 2	95	1190	39
Example 3	125	1250	33
Example 4	190	1230	26
Example 5	280	1080	23
Comparative Example 1	<u>27</u>	610	20
Comparative Example 2	<u>34</u>	700	21
Comparative Example 3	<u>50</u>	860	22
Comparative Example 4	<u>60</u>	960	24
Comparative Example 5	<u>350</u>	790	21
Comparative Example 6	125	450	18

(Comparative Examples 1 to 6)

[0057] For Comparative Examples 1-5, the same process as that of Example 1 was performed except that the melt flow rates of polypropylene resins mixed and kneaded as impregnation resins with the maleic anhydride polypropylene resin were 3.5 g/10 min (Comparative Example 1), 12 g/10 min (Comparative Example 2), 30 g/10 min (Comparative Example 3), 45 g/10 min (Comparative Example 4), and 400 g/10 min (Comparative Example 5).

[0058] For Comparative Example 6, the same process as that of Example 3 was performed except that non-finished glass fibers were used.

[0059] The melt flow rate of each of the polypropylene resins provided as master batch matrix resins in Comparative Examples 1 to 5 was outside of the range of 70 to 300 g/10 min. Accordingly, the glass fibers were not uniformly dispersed, and both the tensile strength (kg/cm²) and the Izod impact strength (kg-cm/cm) of the moldings of Comparative Examples 1 to 6 were much smaller than those of Examples 1 to 5.

[0060] In Comparative Example 6, wettability of the unfinished glass fibers with polypropylene resin was low. Hence, the glass fibers were not suitably impregnated with the polypropylene resin, causing tensile and Izod impact strength reductions.

(Examples 6 to 10, Comparative Example 7)

[0061] For Examples 6-10 and Comparative Example 7, the same experiment as that of Example 3 was conducted except that the impregnation molten resin composition was selected so that maleic anhydride polypropylene resin/polypropylene resin ratios were, by weight, 0/100 (Comparative Example 7), 2/98 (Example 6), 5/95 (Example 7), 10/90 (Example 8), 50/50 (Example 9) and 0/100 (Example 10). Table 2 shows the results of this experiment.

Table 2

	Maleic Anhydride Content in Master Batch Matrix Resin (wt%)	40wt% Glass Fiber Reinforced Molding after Dilution	
		Tensile Strength (kg/cm ²)	Izod Impact Strength (kg·cm/cm)
Example 6	0.024	1230	27
Example 7	0.06	1240	27
Example 8	0.12	1250	27
Example 9	0.60	1250	27
Example 10	1.2	1240	27
Comparative Example 7	0	580	20

[0062] In Comparative Example 7, no chemical bonding between polypropylene and the coupling agent on the glass fiber surface occurred since polypropylene containing no maleic anhydride was used. Consequently, the resulting tensile strength and impact strength were small.

(Comparative Examples 8, 9, Examples 11 to 14)

[0063] In Comparative Example 8, a combination of 40 wt% of chopped strands obtained by cutting to 3 mm the glass rovings used in Example 3, 12 wt% of the modified polypropylene resin used in Example 3, and 48 wt% of the polypropylene resin used in Example 3 having a melt flow rate of 120 g/10 min was mixed and kneaded at 270°C and extruded by a double screw extruder to obtain cylindrical pellets having a length of 10 mm and a diameter of 3 mm. The pellets were injection-molded and physical characteristics of the moldings were measured in the same manner as Example 3. Additionally, the injection moldings were incinerated in a muffle furnace, and the average glass fiber length was determined.

[0064] The same process as that of Example 3 was conducted except that the length of the long-glass fiber reinforced polypropylene pellets were cut to 1 mm (Comparative Example 9), to 2 mm (Example 11), to 5 mm (Example 12), to 30 mm (Example 13), and to 50 mm (Example 14). The average length of glass fibers in the resulting injection moldings was also determined. Table 3 shows the result of this experiment.

Table 3

	Average Length of Glass Fibers in Injection Molding (mm)	40wt% Glass Fiber Reinforced Molding after Dilution	
		Tensile Strength (kg/cm ²)	Izod Impact Strength (kg·cm/cm)
Example 11	0.98	1220	20
Example 12	1.2	1240	25
Example 13	5.0	1250	27
Example 14	8.2	1250	28
Comparative Example 8	0.25	720	9
Comparative Example 9	0.52	960	12

[0065] In Comparative Example 8, the glass fibers were cut during kneading and the resulting shortened fiber length caused a reduction in strength. In Comparative Example 9, the pellet length of 1 mm was half that used in the invention, resulting in tensile strength reduction and a sharp decline in impact strength.

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(Examples 15 to 17, Comparative Examples 10 to 12)

5 **[0066]** For Examples 15-17 and Comparative Examples 10-12, the mixture ratio of the long fiber reinforced polypropylene resin used in Example 3 and a propylene-EPDM copolymer containing 30 wt% of EPDM (melt flow rate: 10 g/10 min) was varied, while the injection moldings and subsequent physical property evaluations were performed in the same manner as Example 3. Table 4 shows the results of this experiment.

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Table 4

	Mixing Ratio (Weight)		Glass Fiber Content in Molding (wt%)	Molding after Dilution	
	Master Batch	Propylene- EPDM Copolymer		Tensile Strength (kg/cm ²)	Izod Impact Strength (kg·cm/cm)
Example 15	10	90	7.5	580	60
Example 16	50	50	37.5	1120	55
Example 17	70	30	49	1180	48
Comparative Example 10	<u>3</u>	<u>97</u>	2.3	220	62
Comparative Example 11	<u>80</u>	<u>20</u>	60	1150	24
Comparative Example 12	<u>100</u>	<u>0</u>	75.0	750	22

[0067] In Comparative Example 10, low tensile strength resulted from a glass fiber content so small that the long fiber reinforcement effect was insufficient. Conversely, in Comparative Examples 11 and 12, the proportion of the master batch in the mixture was so large that tensile strength was lost and a dramatic reduction in impact strength occurred.

[0068] In the prior art, moldings obtained by melting and molding a mixture of a long fiber reinforced polypropylene resin having a large glass fiber content and another polypropylene resin created non-uniform glass fiber dispersion and a resulting strength deficiency. According to the present invention, a mixture of a long glass fiber reinforced polypropylene master batch and a polypropylene resin is prepared in such a manner that glass fibers are treated with a coupling agent, and a modified polypropylene resin having a very low viscosity is used as a matrix resin in the master batch. As a result, uniform glass fiber dispersion occurs even when the master batch has a large glass fiber content, thereby making it possible to form a strong molding having an improved external appearance. Thus, in accordance with the present invention, many types of long glass fiber reinforced moldings serving various uses and purposes can be produced easily and at a low cost from only one long glass fiber reinforced polypropylene master batch having a large glass fiber content by changing the kind of diluting polypropylene resin or by changing the mixture ratio.

Claims

1. A mixture for producing melt process moldings, the mixture comprising:

from 5 to 70 wt% of a master batch of pellets comprising continuous reinforcement glass fibres, the pellets having a length of 2 to 50 mm in the direction along the fibres and the glass fibres in the pellets extending uniformly in parallel with each other through a distance substantially equal to the length of the pellets, and from 30 to 95 wt% of polypropylene resin,

characterised in that the glass fibres are surface-finished with a finishing agent containing a coupling agent and impregnated with a modified polypropylene resin bonded to the coupling agent and having a melt flow rate of from 70g/10min to less than 300 g/10min (ASTM D-1238, load: 2.16 kg, temperature: 230°C).

2. The mixture according to claim 1 wherein the pellets have a glass fibre content of from 60 to 90 wt%.

3. The mixture according to claim 1, or 2 wherein the coupling agent comprises at least one material selected from the group consisting of silane coupling agents and titanate coupling agents.

4. The mixture according to claim 1, 2 or 3 wherein the modified polypropylene resin comprises at least one material selected from the group consisting of carboxylic acid-modified polypropylene resins, acid anhydride-modified polypropylene resins and epoxy-modified polypropylene resins.

5. The mixture according to claim 1, 2, 3 or 4 wherein the modified polypropylene resin is grafted with from 0.01 to 5 wt% of modification monomer.

6. The mixture according to any one of the preceding claims wherein the modified polypropylene resin comprises a trunk polymer having as a unit constituent at least one material selected from the group consisting of: propylene homopolymer, and copolymers of 30 wt% or less of a vinyl-group containing monomer copolymerizable with propylene and 70 wt% or more of propylene.

7. The mixture according to any one of the preceding claims, wherein the coupling agent comprises an aminosilane coupling agent, and the modified polypropylene resin comprises at least one material selected from the group consisting of: polypropylene homopolymer modified with from 0.02 to 1.5 wt% of maleic anhydride, and ethylene-propylene copolymers modified with from 0.02 to 1.5 wt% of maleic anhydride and having an ethylene content of 5 wt% or less.

8. The mixture according to any one of the preceding claims, wherein the melt flow rate of the modified polypropylene resin measured by ASTM method D-1238 (load: 2.16 kg, temperature: 230°C) is within the range of from 80 g/10 min to 150 g/10 min.

9. The mixture according to any one of the preceding claims, wherein the glass fibre content of the pellets is within the range of from 60 to 90 wt%, preferably from 70 to 85 wt%.

10. The mixture according to any one of the preceding claims, wherein the length of the pellets is within the range of from 4 to 15 mm.
- 5 11. The mixture according to any one of the preceding claims, wherein the polypropylene resin mixed with the pellets has a melt flow rate within the range of from 5 g/10 min to 150 g/10 min as measured by ASTM method D-1238 (load: 2.16 kg, temperature: 230°C).
- 10 12. The mixture according to any one of the preceding claims, wherein the polypropylene resin mixed with the pellets comprises at least one material selected from the group consisting of: propylene homopolymer, and copolymers of 30 wt% or less of a vinyl-group containing monomer copolymerizable with propylene and 70 wt% or more of propylene.
- 15 13. The mixture according to claim 12, wherein the copolymer of propylene and a vinyl-group containing monomer copolymerizable with propylene comprises at least one material selected from the group consisting of: ethylene-propylene block copolymer having an ethylene content of from 10 to 30 wt%, EPDM-propylene copolymer having an EPDM content of from 10 to 30 wt%, and a mixture of the ethylene-propylene block copolymer and the EPDM-propylene copolymer.
- 20 14. The mixture according to any one of the preceding claims, wherein the content of the pellets is limited within the range of from 20 to 50 wt% while the content of the polypropylene resin mixed with the pellets is limited within the range of from 80 to 50 wt%.
- 25 15. A melt process molding obtained by melting and molding a melt process molding mixture as claimed in any preceding claim.
16. A melt process molding as claimed in claim 15 wherein the glass fibres have an average length of from 0.8 to 10 mm.
17. A method of preparing a mixture for producing melt process moldings comprising:
30 forming glass fibres into a bundle;
impregnating the bundle with a modified polypropylene resin having a functional group capable of chemically bonding with a coupling agent;
dividing the resulting impregnated bundle into a multiplicity of pellets; and
35 mixing the pellets with polypropylene resin,
characterised in that the glass fibres are coated with a finishing agent containing a coupling agent before they are formed into a bundle, and in that the modified polypropylene resin has a melt flow rate of from 70g/10min to less than 300g/10min (ASTM D-1238, load : 2.16 kg, temperature 230°C).
- 40 18. A method of preparing a mixture for producing melt process moldings comprising:
impregnating a bundle of glass fibres with a modified polypropylene resin having a functional group capable of chemically bonding with a coupling agent;
dividing the resulting impregnated bundle into a multiplicity of pellets; and
45 mixing the pellets with polypropylene resin,
characterised in that the method includes the step of coating individual glass fibres with a finishing agent containing a coupling agent, and in that the modified polypropylene resin has a melt flow rate of from 70g/10min to less than 300g/10min (ASTM D-1238, load : 2.16 kg, temperature 230°C).
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Patentansprüche

- 55 1. Mischung zum regulinischen Herstellen von Formteilen mit 5 bis 70 % einer ununterbrochene, verstärkende Glasfasern enthaltenden Grundmischung aus Pellets, die eine Länge von 2 bis 50 mm in der Ausdehnungsrichtung der Glasfasern aufweisen, welche sich gleichmäßig parallel zueinander in einer Länge erstrecken, die im wesentlichen mit der Gesamtlänge der Pellets übereinstimmt, und 30 bis 95 % eines Polypropylenharzes, **dadurch gekennzeichnet**, daß die Glasfasern mit einem ein Kopplungsmittel enthaltenden Appreturmittel oberflächenbehan-

delt und mit einem modifizierten Polypropylenharz imprägniert sind, das an das Kopplungsmittel gebunden ist und einen Schmelzindex von 70 g/10 min bis weniger als 300 g/10 min (ASTM D-1238, Gewicht: 2,16 kg, Temperatur: 230 °C) aufweist.

- 5 2. Mischung nach Anspruch 1, **dadurch gekennzeichnet**, daß die Pellets einen Glasfasergehalt von 60 bis 90 % aufweisen.
3. Mischung nach Anspruch 1 oder 2, **dadurch gekennzeichnet**, daß das Kopplungsmittel mindestens einen Bestandteil aus der Gruppe der Silan- und Titanat-Kopplungsmittel enthält.
- 10 4. Mischung nach einem der Ansprüche 1 bis 3, **dadurch gekennzeichnet**, daß das Polypropylenharz mindestens einen Bestandteil aus der Gruppe der mit Carboxylsäuren, Anhydriden oder mit Epoxiden modifizierten Polypropylenharze enthält.
- 15 5. Mischung nach einem der Ansprüche 1 bis 4, **dadurch gekennzeichnet**, daß das modifizierte Polypropylenharz zusammen mit 0,01 bis 5 % des Modifikationsmittels pfpöpolymerisierte.
6. Mischung nach einem der Ansprüche 1 bis 5, **dadurch gekennzeichnet**, daß das modifizierte Polypropylenharz eine Basis aus einem Propylenhomopolymer und/oder Copolymeren mit mindestens 70 % Propylen und bis 30 % mit Polypropylen polymerisierbare Monomere enthaltenden Vinylgruppen enthält.
- 20 7. Mischung aus einem der Ansprüche 1 bis 6, **dadurch gekennzeichnet**, daß das Kopplungsmittel ein Amino-Silan und das modifizierte Polypropylen ein Propylenhomopolymer modifiziert mit 0,02 bis 1,5 % Maleinsäureanhydrid und/oder ein Ethylen-Propylen-Copolymer modifiziert mit 0,02 bis 1,5 % Maleinsäureanhydrid und mit einem Ethylengehalt bis zu 5 % enthält.
- 25 8. Mischung nach einem der Ansprüche 1 bis 7, **dadurch gekennzeichnet**, daß der Schmelzindex des modifizierten Polypropylens 80 bis 150 g/10 min nach ASTM D-1238 (Gewicht: 2,16 kg, Temperatur: 230 °C) beträgt.
- 30 9. Mischung nach einem der Ansprüche 1 bis 8, **dadurch gekennzeichnet**, daß der Glasfasergehalt des Pellets 60 bis 90 %, vorzugsweise 70 bis 85 % beträgt.
10. Mischung nach einem der Ansprüche 1 bis 9, **dadurch gekennzeichnet**, daß die Pellets eine Länge von 4 bis 15 mm aufweisen.
- 35 11. Mischung nach einem der Ansprüche 1 bis 10, **dadurch gekennzeichnet**, daß das mit den Pellets gemischte Polypropylenharz einen Schmelzindex von 5 bis 150 g/10 min nach ASTM D-1238 (Gewicht: 2,16 kg, Temperatur: 230 °C) besitzt.
- 40 12. Mischung nach einem der Ansprüche 1 bis 11, **dadurch gekennzeichnet**, daß das mit den Pellets gemischte Polypropylenharz ein Propylenhomopolymer und/oder Copolymere mit mindestens 70 % Propylen und bis 30 % mit Polypropylen polymerisierbare Monomere enthaltenden Vinylgruppen enthält.
- 45 13. Mischung nach Anspruch 12, **dadurch gekennzeichnet**, daß das Copolymer aus Propylen und Vinylgruppen, die mit Polypropylen polymerisierbaren Monomere enthalten, ein Ethylen-Propylen-Polymer mit einem Ethylengehalt von 10 bis 30 % und/oder ein EPDM Propylen-Copolymer mit einem EPDM-Gehalt von 10 bis 30 % enthält.
14. Mischung nach einem der Ansprüche 1 bis 13, **gekennzeichnet durch** 20 bis 50 % Pellets und 80 bis 50 % Polypropylenharz.
- 50 15. Formteil, das durch Schmelzen und Formen einer Mischung nach einem der Ansprüche 1 bis 14 erzeugt wurde.
16. Formteil nach Anspruch 15, **gekennzeichnet durch** eine durchschnittliche Länge der Glasfasern von 0,8 bis 10 mm.
- 55 17. Verfahren zum Herstellen einer Mischung bei der Fertigung von Formteilen durch Schmelzen, bei dem die Glasfasern gebündelt, das Bündel mit einem modifizierten Polypropylenharz mit einer ein Kopplungsmittel bindenden funktionellen Gruppe getränkt, das imprägnierte Bündel in Pellets unterteilt wird und die Pellets mit einem Poly-

propylenharz gemischt werden, **dadurch gekennzeichnet**, daß die Glasfasern vor dem Bündeln mit einem ein Kopplungsmittel enthaltenden Appreturmittel überzogen werden, und das Polypropylenharz einen Schmelzindex von 70 bis weniger als 300 g/10 min aufweist (ASTM D-1238, Gewicht: 2,16 kg; Temperatur: 230 °C).

- 5 18. Verfahren zum Herstellen von Mischungen bei der Fertigung von Formteilen durch Schmelzen, bei dem das Bündel mit einem modifizierten Polypropylenharz mit einer ein Kopplungsmittel bindenden funktionellen Gruppe getränkt, das imprägnierte Bündel in Pellets unterteilt wird und die Pellets mit Polypropylen gemischt werden, **dadurch gekennzeichnet**, daß die einzelnen Glasfasern mit einem ein Kopplungsmittel enthaltenden Appreturmittel über-
10 zogen werden, und das Polypropylenharz einen Schmelzindex von 70 bis weniger als 300 g/10 min aufweist (ASTM D-1238, Gewicht: 2,16 kg; Temperatur: 230 °C).

Revendications

- 15 1. Mélange permettant de produire des pièces moulées en fusion, le mélange comprenant :

5 à 70 % en poids d'un mélange-maître de granulés comprenant des fibres de verre de renforcement continues, les granulés ayant une longueur de 2 à 50 mm dans la direction longitudinale des fibres et les fibres de verre dans les granulés s'étendant de manière homogène parallèlement les unes aux autres sur une distance sensiblement égale à la longueur des granulés, et
20 30 à 95 % en poids de résine de polypropylène,

caractérisé en ce que les fibres de verre sont soumises à une finition de surface au moyen d'un agent de finition contenant un agent de pontage et imprégnées d'une résine de polypropylène modifiée, liée à l'agent de pontage et présentant un indice de fusion compris entre 70g/10 min. et 300 g/10 min. exclus (ASTM D-1238, charge : 2,16 kg, température : 230°C).

2. Mélange selon la revendication 1, dans lequel les granulés contiennent de 60 à 90 % en poids de fibres de verre.

- 30 3. Mélange selon la revendication 1 ou 2, dans lequel l'agent de pontage comprend au moins un composant choisi parmi le groupe constitué par les agents de pontage à base de silane et les agents de pontage à base de titane.

4. Mélange selon la revendication 1, 2 ou 3, dans lequel la résine de polypropylène modifiée comprend au moins un composant choisi parmi le groupe constitué par les résines de polypropylène modifiées par l'acide carboxylique, les résines de polypropylène modifiées par un anhydride d'acide et les résines de polypropylène modifiées par l'époxy.
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5. Mélange selon la revendication 1, 2, 3 ou 4, dans lequel la résine de polypropylène modifiée est greffée avec 0,01 à 5 % en poids d'un monomère de modification.
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6. Mélange selon l'une quelconque des revendications précédentes, dans lequel la résine de polypropylène modifiée comprend un polymère de base possédant, comme constituant élémentaire, au moins un composant choisi parmi le groupe constitué par : un homopolymère de propylène ; et des copolymères contenant au maximum 30 % en poids d'un monomère contenant un groupe vinyle, copolymérisable avec le propylène, et 70 % en poids ou plus de propylène.
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7. Mélange selon l'une quelconque des revendications précédentes, dans lequel l'agent de finition comprend un agent de pontage à base d'aminosilane et la résine de polypropylène modifiée comprend au moins un composant choisi parmi le groupe constitué par : un homopolymère de polypropylène modifié par 0,02 à 1,5 % en poids d'anhydride maléique et des copolymères éthylène-propylène modifiés par 0,02 à 1,5 % en poids d'anhydride maléique et présentant une teneur en éthylène inférieure ou égale à 5 % en poids.
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8. Mélange selon l'une quelconque des revendications précédentes, dans lequel l'indice de fusion de la résine de polypropylène modifiée, mesuré selon la méthode ASTM D-1238 (charge 2,16 kg, température : 230 °C) est compris entre 80 g/10 min. et 150 g/10 min.
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9. Mélange selon l'une quelconque des revendications précédentes, dans lequel la teneur en fibres de verre des granulés est comprise entre 60 et 90 % en poids, de préférence entre 70 et 85 % en poids.

10. Mélange selon l'une quelconque des revendications précédentes, dans lequel la longueur des granulés est comprise entre 4 et 15 mm.
- 5 11. Mélange selon l'une quelconque des revendications précédentes, dans lequel la résine de polypropylène mélangée aux granulés présente un indice de fusion compris entre 5 g/10 min. et 150 g/10min. mesuré selon la méthode ASTM D-1238 (charge 2,16 kg, température : 230 °C).
- 10 12. Mélange selon l'une quelconque des revendications précédentes, dans lequel la résine de polypropylène mélangée aux granulés comprend au moins un composant choisi parmi le groupe constitué par : un homopolymère de propylène, et des copolymères contenant au maximum 30 % en poids d'un monomère contenant un groupe vinyle, copolymérisable avec le propylène ; et 70 % en poids ou plus de propylène.
- 15 13. Mélange selon la revendication 12, dans lequel le copolymère de propylène et d'un monomère contenant un groupe vinyle, copolymérisable avec le propylène, comprend au moins un composant choisi parmi le groupe constitué par : un copolymère en masse éthylène-propylène ayant une teneur en éthylène comprise entre 10 et 30 % en poids, un copolymère EPDM-propylène ayant une teneur en EPDM comprise entre 10 et 30 % en poids et un mélange du copolymère en masse éthylène-propylène et du copolymère EPDM-propylène.
- 20 14. Mélange selon l'une quelconque des revendications précédentes, dans lequel la teneur en granulés est limitée à la plage comprise entre 20 et 50 % en poids tandis que la teneur en résine de polypropylène mélangée aux granulés est limitée à la plage comprise entre 80 et 50 % en poids.
- 25 15. Pièce moulée en fusion, obtenue par fusion et moulage d'un mélange de moulage en fusion selon l'une quelconque des revendications précédentes.
16. Pièce moulée en fusion selon la revendication 15, dans laquelle les fibres de verre ont une longueur moyenne comprise entre 0,8 et 10 mm.
- 30 17. Méthode de préparation d'un mélange destiné à la production de pièces moulées en fusion, comprenant :
la formation d'un faisceau de fibres de verre ;
l'imprégnation du faisceau avec une résine de polypropylène modifiée ayant un groupe fonctionnel capable de se lier chimiquement avec un agent de pontage ;
la division du faisceau imprégné résultant en une multitude de granulés ; et
35 le mélange des granulés avec de la résine de polypropylène, caractérisée en ce que les fibres de verre sont recouvertes d'un agent de finition contenant un agent de pontage avant d'être rassemblées en faisceau, et en ce que la résine de polypropylène modifiée présente un indice de fusion compris entre 70 g/10 min. et 300 g/10 min. exclus (ASTM D-1238, charge : 2,16 kg, température : 230 °C).
- 40 18. Méthode de préparation d'un mélange destiné à la production de pièces moulées en fusion, comprenant :
l'imprégnation d'un faisceau de fibres de verre avec une résine de polypropylène modifiée ayant un groupe fonctionnel capable de se lier chimiquement avec un agent de pontage ;
la division du faisceau imprégné résultant en une multitude de granulés ; et
45 le mélange des granulés avec de la résine de polypropylène, caractérisée en ce que la méthode inclut l'étape de revêtement de chaque fibre de verre avec un agent de finition contenant un agent de pontage, et en ce que la résine de polypropylène modifiée présente un indice de fusion compris entre 70 g/10 min. et 300 g/10 min. exclus (ASTM D-1238, charge : 2,16 kg, température : 230 °C).
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